



J007 Rec'd PCT/PTO 07 JAN 2002

FORM PTO-1390 U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371 <i>JAN 07 2002</i>		ATTORNEY'S DOCKET NUMBER: 3501-1001
		U.S. PPLN. NO. (Unknown, see 37 CFR 1.5)
INTERNATIONAL APPLICATION NO.: PCT/FI00/00033		INTERNATIONAL FILING DATE: 4 JULY 2000
		PRIORITY DATE CLAIMED: 5 JULY 1999
TITLE OF INVENTION: COMPOSITION TO BE USED IN PAINTS		
APPLICANT(S) FOR DO/EO/US: Mika PERÄLÄ and Seppo TIKKANEN		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
1.	<input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.	
2.	<input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.	
3.	<input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).	
4.	<input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.	
5.	<input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ul style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau. (see attached copy of PCT/IB/308)</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ul>	
6.	<input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).	
7.	<input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)). <ul style="list-style-type: none"> <li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ul>	
8.	<input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).	
9.	<input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).	
10.	<input type="checkbox"/> A translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).	
Item 11. to 16. below concern document(s) or information included:		
11.	<input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.	
12.	<input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.	
13.	<input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment.	
14.	<input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.	
15.	<input type="checkbox"/> A substitute specification.	
16.	<input checked="" type="checkbox"/> Other items or information:	
INTERNATIONAL PRELIMINARY EXAMINATION REPORT (PCT/IPEA/409), INTERNATIONAL PUBLICATION, INTERNATIONAL SEARCH REPORT (PCT/ISA/210), FINNISH SEARCH REPORT, PCT/IB/308, ABSTRACT on a separate sheet, APPLICATION DATA SHEET		

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)		INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NO.																																																																																									
<b>10/019962</b>		PCT/F100/00613	3501-1001																																																																																									
CALCULATIONS PTO USE ONLY																																																																																												
17. <input checked="" type="checkbox"/> The following fees are submitted:																																																																																												
<b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b>																																																																																												
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$ 1,040.00																																																																																												
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$ 890.00																																																																																												
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$ 740.00																																																																																												
International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$ 710.00																																																																																												
International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$ 100.00																																																																																												
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b> \$ 1,040.00																																																																																												
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492(e)).																																																																																												
<table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th>CLAIMS</th> <th>NUMBER FILED</th> <th>NUMBER EXTRA</th> <th>RATE</th> <th>\$</th> </tr> </thead> <tbody> <tr> <td>Total claims</td> <td>12 - 20 =</td> <td>0</td> <td>X \$18.00</td> <td>\$</td> </tr> <tr> <td>Independent claims</td> <td>1 - 3 =</td> <td>0</td> <td>X \$84.00</td> <td>\$</td> </tr> <tr> <td colspan="3"><b>MULTIPLE DEPENDENT CLAIMS(S) (if applicable)</b></td> <td>+ \$280.00</td> <td>\$</td> </tr> <tr> <td colspan="4"><b>TOTAL OF ABOVE CALCULATIONS =</b></td> <td>\$ 1,040.00</td> </tr> <tr> <td colspan="4">Reduction of <math>\frac{1}{2}</math>, if applicant is entitled to Small Entity status under 37 CFR 1.27.</td> <td>+ \$ 520.00</td> </tr> <tr> <td colspan="4"></td> <td><b>SUBTOTAL =</b> \$ 520.00</td> </tr> <tr> <td colspan="4">Processing fee of \$130 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492(f)).</td> <td>\$</td> </tr> <tr> <td colspan="4"></td> <td><b>TOTAL NATIONAL FEE =</b> \$ 520.00</td> </tr> <tr> <td colspan="4">Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property</td> <td>+ \$ 40.00</td> </tr> <tr> <td colspan="4"></td> <td><b>TOTAL FEES ENCLOSED =</b> \$ 560.00</td> </tr> <tr> <td colspan="4"></td> <td>Amount to be refunded:</td> </tr> <tr> <td colspan="4"></td> <td>charged:</td> </tr> <tr> <td>a. <input checked="" type="checkbox"/></td> <td colspan="3">A check in the amount of \$ <u>560.00</u> to cover the above fees is enclosed.</td> </tr> <tr> <td>b. <input type="checkbox"/></td> <td colspan="3">Please charge my Deposit Account No. <b>25-0120</b> in the amount of \$ to cover the above fees. A duplicate copy of this sheet is enclosed.</td> </tr> <tr> <td>c. <input checked="" type="checkbox"/></td> <td colspan="3">The Commissioner is hereby authorized to charge any additional fees which may be required by 37 CFR 1.16 and 1.17, or credit any overpayment to Deposit Account No. <b>25-0120</b>. A duplicate copy of this sheet is enclosed.</td> </tr> <tr> <td colspan="4">SEND ALL CORRESPONDENCE TO:</td> </tr> <tr> <td colspan="2"> <b>CUSTOMER NO. 00466</b>            YOUNG &amp; THOMPSON            745 South 23rd Street            2nd Floor            Arlington, VA 22202            (703) 521-2297            facsimile (703) 685-0573         </td> <td colspan="2">   <b>00466</b>  <small>PATENT TRADEMARK OFFICE</small>            7 JANUARY 2002         </td> </tr> <tr> <td colspan="4" style="text-align: right;">           By             Benoit Castel            Attorney for Applicants            Registration No. 35,041         </td> </tr> </tbody></table>				CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	Total claims	12 - 20 =	0	X \$18.00	\$	Independent claims	1 - 3 =	0	X \$84.00	\$	<b>MULTIPLE DEPENDENT CLAIMS(S) (if applicable)</b>			+ \$280.00	\$	<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 1,040.00	Reduction of $\frac{1}{2}$ , if applicant is entitled to Small Entity status under 37 CFR 1.27.				+ \$ 520.00					<b>SUBTOTAL =</b> \$ 520.00	Processing fee of \$130 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492(f)).				\$					<b>TOTAL NATIONAL FEE =</b> \$ 520.00	Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				+ \$ 40.00					<b>TOTAL FEES ENCLOSED =</b> \$ 560.00					Amount to be refunded:					charged:	a. <input checked="" type="checkbox"/>	A check in the amount of \$ <u>560.00</u> to cover the above fees is enclosed.			b. <input type="checkbox"/>	Please charge my Deposit Account No. <b>25-0120</b> in the amount of \$ to cover the above fees. A duplicate copy of this sheet is enclosed.			c. <input checked="" type="checkbox"/>	The Commissioner is hereby authorized to charge any additional fees which may be required by 37 CFR 1.16 and 1.17, or credit any overpayment to Deposit Account No. <b>25-0120</b> . A duplicate copy of this sheet is enclosed.			SEND ALL CORRESPONDENCE TO:				<b>CUSTOMER NO. 00466</b> YOUNG & THOMPSON 745 South 23rd Street 2nd Floor Arlington, VA 22202 (703) 521-2297 facsimile (703) 685-0573		 <b>00466</b> <small>PATENT TRADEMARK OFFICE</small> 7 JANUARY 2002		By  Benoit Castel Attorney for Applicants Registration No. 35,041			
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$																																																																																								
Total claims	12 - 20 =	0	X \$18.00	\$																																																																																								
Independent claims	1 - 3 =	0	X \$84.00	\$																																																																																								
<b>MULTIPLE DEPENDENT CLAIMS(S) (if applicable)</b>			+ \$280.00	\$																																																																																								
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 1,040.00																																																																																								
Reduction of $\frac{1}{2}$ , if applicant is entitled to Small Entity status under 37 CFR 1.27.				+ \$ 520.00																																																																																								
				<b>SUBTOTAL =</b> \$ 520.00																																																																																								
Processing fee of \$130 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492(f)).				\$																																																																																								
				<b>TOTAL NATIONAL FEE =</b> \$ 520.00																																																																																								
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				+ \$ 40.00																																																																																								
				<b>TOTAL FEES ENCLOSED =</b> \$ 560.00																																																																																								
				Amount to be refunded:																																																																																								
				charged:																																																																																								
a. <input checked="" type="checkbox"/>	A check in the amount of \$ <u>560.00</u> to cover the above fees is enclosed.																																																																																											
b. <input type="checkbox"/>	Please charge my Deposit Account No. <b>25-0120</b> in the amount of \$ to cover the above fees. A duplicate copy of this sheet is enclosed.																																																																																											
c. <input checked="" type="checkbox"/>	The Commissioner is hereby authorized to charge any additional fees which may be required by 37 CFR 1.16 and 1.17, or credit any overpayment to Deposit Account No. <b>25-0120</b> . A duplicate copy of this sheet is enclosed.																																																																																											
SEND ALL CORRESPONDENCE TO:																																																																																												
<b>CUSTOMER NO. 00466</b> YOUNG & THOMPSON 745 South 23rd Street 2nd Floor Arlington, VA 22202 (703) 521-2297 facsimile (703) 685-0573		 <b>00466</b> <small>PATENT TRADEMARK OFFICE</small> 7 JANUARY 2002																																																																																										
By  Benoit Castel Attorney for Applicants Registration No. 35,041																																																																																												

10/019962

JC13 Rec'd PCT/PTO 07 JAN 2002

PATENT  
3501-1001

**IN THE U.S. PATENT AND TRADEMARK OFFICE**

In re application of: Mika PERÄLÄ et al.  
Appl. No.: (unassigned)  
Filed: January 7, 2002  
For: COMPOSITION TO BE  
USED IN PAINTS

**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Washington, DC 20231

January 7, 2002

Sir:

The following preliminary amendments and remarks are respectfully submitted in connection with the above-identified application.

**IN THE ABSTRACT OF THE DISCLOSURE:**

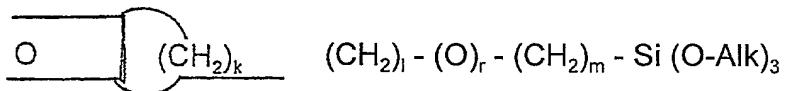
Please replace the Abstract of the Disclosure with the rewritten Abstract of the Disclosure attached hereto.

**IN THE CLAIMS:**

Please amend the claims as follows:

--3. (amended) A composition as claimed in claim 1,  
**characterized** in that the non-aromatic epoxy resin is a branched aliphatic  
epoxy resin.--

--9. (amended) A composition as claimed in claim 1, **characterized** in that the epoxy silane has the formula



where k is an integer between 0 and 4, r is 0 or 1, l is an integer between 1 and 6, m is an integer between 1 and 6 and Alk is an alkyl group having 1 to 6 carbon atoms.--

--12. (amended) A kit, **characterized** by comprising a container A, which contains a composition according to claim 1, and a container B, which contains a hardener, whereby the container A and/or B may further contain conventional additives.--

REMARKS

The above changes in the claims merely place this national phase application in the same condition as it was during Chapter II of the international phase, with the multiple dependencies being removed.

Claims 1-12 are pending in the present application.

Entry of the above amendments is earnestly solicited. An early and favorable first action on the merits is earnestly requested.

Should there be any matters that need to be resolved in the present application, the Examiner is respectfully requested to contact the undersigned at the telephone number listed below.

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached page is captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE."

The Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 25-0120 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17.

Respectfully submitted,

YOUNG & THOMPSON

Benoit Castel

---

Benoit Castel, Reg. No. 35,041  
745 South 23<sup>rd</sup> Street  
Arlington, VA 22202  
Telephone (703) 521-2297

BC/lmt  
Attachments

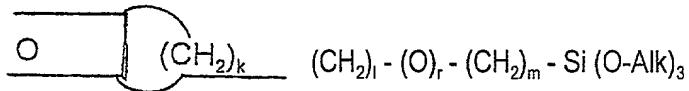
VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS:

The claims have been amended as follows:

--3. (amended) A composition as claimed in claim 1 or 2, **characterized** in that the non-aromatic epoxy resin is a branched aliphatic epoxy resin.--

--9. (amended) A composition as claimed in ~~any one of claims~~ claim 1 to 8, **characterized** in that the epoxy silane has the formula



where k is an integer between 0 and 4, r is 0 or 1, l is an integer between 1 and 6, m is an integer between 1 and 6 and Alk is an alkyl group having 1 to 6 carbon atoms.--

--12. (amended) A kit, **characterized** by comprising a container A, which contains a composition according to ~~any one of claims~~ claim 1 to 11, and a container B, which contains a hardener, whereby the container A and/or B may further contain conventional additives.--

## ABSTRACT

The invention relates to a

A paint composition comprising has a resin constituent which includes (i) a non-aromatic epoxy resin, (ii) a polysiloxane and (iii) an epoxysilane. The paint composition of the invention has an anti-corrosive effect.

IPR/1

1

## COMPOSITION TO BE USED IN PAINTS

### FIELD OF THE INVENTION

The present invention relates to an environmentally friendly, epoxy-resin-based anti-corrosive composition to be used in paints, which composition has high dry solids content and low viscosity and which can be applied to protect steel structures and/or concrete surfaces.

### BACKGROUND OF THE INVENTION

Epoxy-, alkyd- and polyurethane-based paints and combinations thereof are commonly used to protect steel structures and/or concrete surfaces against corrosion. In general, polyurethane-based paints require a primer. Their greatest disadvantage, however, is related to health factors. The dry solids content of the polyurethane paints is low, which results in a great amount of volatile substances (solvents) and limits the use of said paints. Even though epoxy paints have several good properties, such as good adhesion to steel, concrete and other substrates, good mechanical strength and chemical resistance, it is necessary to use weather-resistant alkyd, or in particular, polyurethane paint for topcoating, because the epoxy paint chalks, i.e. becomes matt, and often yellows when exposed to weather. The required film thicknesses are within the range of about 150 to 500 µm, which often requires 2 to 4 repeated treatments. Even though the epoxy paints have been developed to achieve thicker films with fewer treatments and a slight improvement in the weatherability has also been achieved, the use of polyurethane as a top coat cannot have been avoided so far. The amount of volatile organic content (VOC) still constitutes a problem for both epoxy and polyurethane paints.

High viscosity has also limited the use of previously known epoxy paints. A low ambient temperature has also imposed restrictions on their use in the open air. Quite recently, reduction in viscosity and improvement in weather resistance of the epoxy paints are achieved by modifying epoxy resins with acrylic monomers and oligomers. At the same time, the use of acrylic monomers and oligomers has considerably shortened drying times and exploitation times of the paints.

Further, silicone paints and silicate paints based on organic and inorganic silicon compounds have been known for a long time. Silicone paints resist well heat, up to 600 °C, but they dry poorly and yellow easily. Zinc silicate paints have often been used as a primer for demanding anti-corrosive

paintings.

Polysiloxane-based paints, based on silicon and oxygen compounds, have also been developed having excellent weather resistance and particularly improved UV light resistance even as compared with the polyurethanes. However, due to their high price, these polysiloxanes acting as binders are used as a sole binder in paints only in exceptional cases, such as in space technology where requirements are extremely high. The high price has forced paint manufacturers to use silicone and siloxane compounds instead of polysiloxanes for improving and modifying the properties of other binders.

An epoxy-based polysiloxane coating composition useful for corrosion prevention is known from WO 96/16109, the composition including a non-aromatic hydrogenated epoxy resin, a polysiloxane and an organooxysilane. As preferred non-aromatic epoxide resins are stated hydrogenated cyclohexane dimethanol and diglycidyl ethers of hydrogenated Bisphenol A-type. When the composition dries, the non-aromatic epoxy resin used forms a straight-chain epoxy polymer and not a three-dimensional structure, which is possible in the solution of the present invention. Good weather resistance of the known composition is believed to be specifically due to the linear epoxy polymer. On the other hand, it has been found that the drying times of these compositions are relatively long.

A casting resin composition used for covering optoelectronic components is known from US Patent 5,492,981. Said US patent thus relates to a field of use which is different from the field of use of the present invention. For instance, the hardener type and the curing temperature cited in said patent publication indicate that the known composition in question is not related to paints in any manner whatsoever. The resin component of the casting resin composition according to the US patent contains 5 to 95 % by weight of a condensation product which is an epoxy-groups-containing polysiloxane formed of a silanol and an epoxyalkoxysilane at an elevated temperature. According to the publication, the resin component also contains 5 to 95% by weight of epoxy resin. The solution of the present invention, in turn, uses polysiloxane, which does not include epoxy groups, and separately epoxy silane and aliphatic epoxy resin.

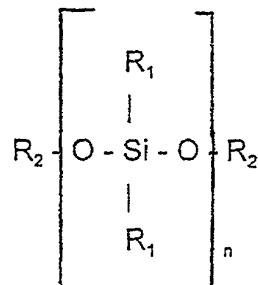
The object of the invention is thus to provide a low-viscosity anti-corrosive paint composition which avoids the problems of the corresponding, known epoxy-based paint compositions. The present invention is based on the

- use of a combination of aliphatic epoxy constituents and specific polysiloxane-containing constituents. Now, it has been found that a paint composition having excellent properties is obtained by using epoxy silane, instead of organo-oxysilane (cf. WO 96/16109), together with an aliphatic epoxy resin. The composition of the invention has an advantage that it has low viscosity, whereby diluting is not generally needed, and that it dries quickly also at low temperatures, even at -5 °C. The drying rate and curing at low temperatures of known epoxy compositions can be improved by specific accelerators, which, however, do not act at the same temperatures or they cause other problems, such as more intense yellowing than conventionally and matting (chalking) of a painted surface. These accelerators can also be toxic, for instance, phenol derivatives.

The compositions of the invention have a further advantage that it is possible to apply new coatings over the paints containing said compositions after a long time without that any adhesion or curling problems occur. They need not be coated with a separate weather-resistant topcoat either, film thicknesses can be reduced in many cases and the intervals between maintenance paintings can be prolonged, which in turn reduces costs.

#### SUMMARY OF THE INVENTION

- The present invention relates to a composition to be used in paints, which is characterized by comprising a resin constituent which includes
- i) a non-aromatic epoxy resin,
  - ii) a polysiloxane having the formula:

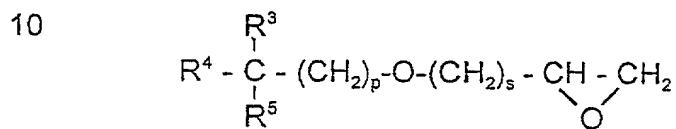


- 30 where  $R^1$  is hydroxyl or an alkyl, aryl or alkoxy group having up to 6 carbon atoms,  $R^2$  is a hydrogen or an alkyl or aryl group having up to 6 carbon atoms and  $n$  is a number selected so that the molar mass of the polysiloxane is within the range of 400 to 2000, and
- iii) an epoxy silane which acts as a crosslinking agent between the epoxy and siloxane chains.

The invention also relates to a kit, which is characterized by comprising a container A, which contains a composition of the present invention, and a container B, which contains a hardening agent, whereby the container A and/or B may further contain conventional additives. The additives may include accelerators, water and other conventional additives.

#### DETAILED DESCRIPTION OF THE INVENTION

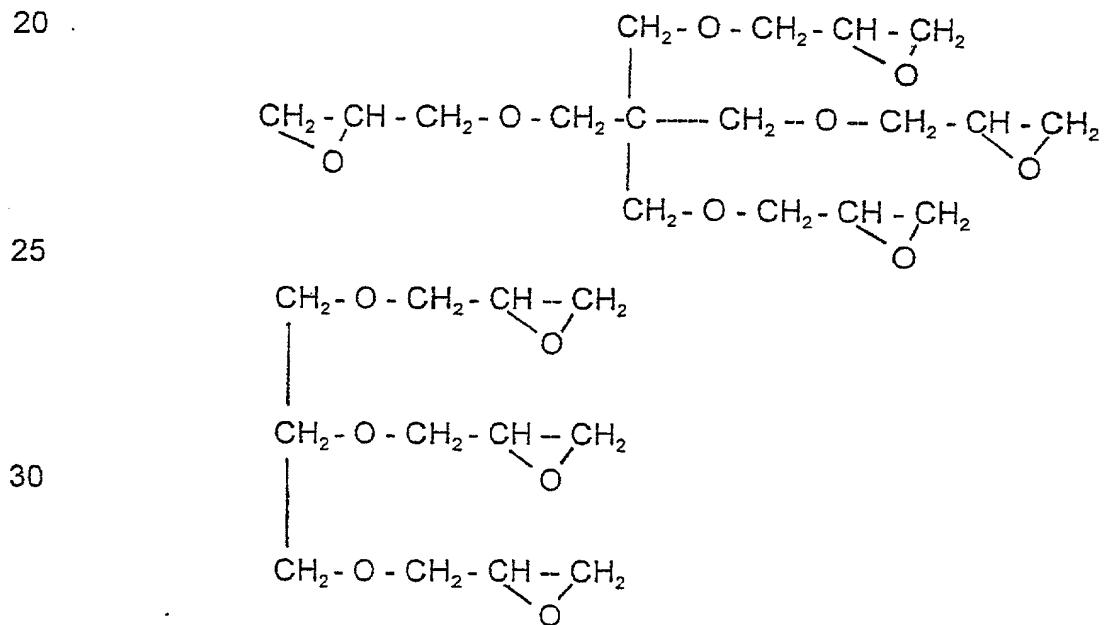
In the composition of the invention, the non-aromatic epoxy resin is preferably an aliphatic branched epoxy resin. A suitable branched aliphatic epoxy resin has the general formula:

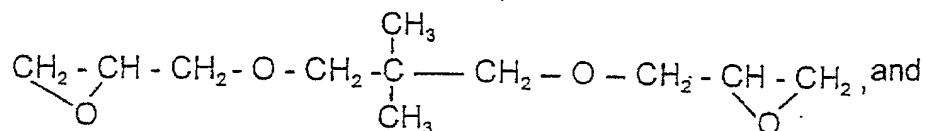


where p is an integer between 0 and 3, s is an integer between 1 and 3, R<sup>3</sup> and R<sup>4</sup> represent independently C<sub>1-6</sub>alkyl or a group -(CH<sub>2</sub>)<sub>p</sub>-O-(CH<sub>2</sub>)<sub>s</sub>-CH-CH<sub>2</sub>,

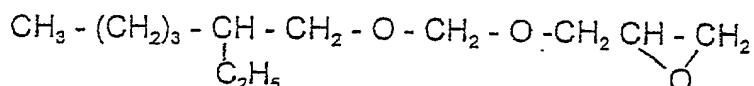
where p and s are as defined above and R<sup>5</sup> is hydrogen, C<sub>1-6</sub>alkyl or a group -(CH<sub>2</sub>)<sub>p</sub>-O-(CH<sub>2</sub>)<sub>s</sub>-CH-CH<sub>2</sub>, where p and s are as defined above.

The preferred branched aliphatic epoxy resins include



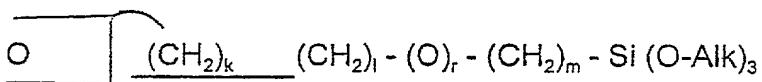


5



10 Polysiloxanes described in WO 96/16109 can be generally used as  
a polysiloxane constituent of the resin component in the compositions of the  
invention. Particularly preferable are substances under commercial names  
DC-3074 and DC-3037 available from the manufacturer Dow Corning, which  
are methoxy functional polysiloxanes. The amount of polysiloxane in the paint  
15 composition is 15 to 45 % by weight.

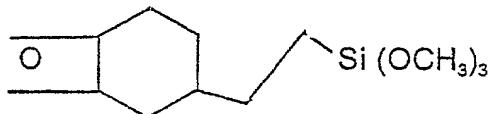
Suitable epoxy silanes include those having the general formula:



where k is an integer between 0 and 4, l is an integer between 1 and 6, r is 0 or 1, m is an integer between 1 and 6 and Alk is an alkyl group having 1 to 6 carbon atoms. Particularly preferable epoxy silanes are glycidoxypropyl-trimethoxysilane having the formula

25 

and  $\beta$ -(3,4-epoxycyclohexyl)-ethyltrimethoxysilane having the formula



30

In the composition of the invention the weight ratio between the epoxy silane, polysiloxane and non-aromatic epoxy resin is suitably 1:2-5:2-5.

In connection with the present invention, conventional hardening agents can be used as hardeners. Such hardeners include, for instance, aliphatic amines, aliphatic amine adducts, polyamidoamines, cycloaliphatic amines and adducts thereof, aromatic amines, Mannich bases and ketimines.

all of which may be substituted wholly or in part with aminosilanes. Amine hardeners useful for the present invention are described in WO 96/16109, for instance.

5      Organic tin compounds, such as dibutyl tin dilaurate (DBTL) or organic salts, for instance naphthanates, of some metals, such as cobalt, zinc, manganese, iron and lead, can be used as an accelerator in the compositions of the invention.

10     Water can be included in the compositions of the invention, either separately added or in the form of water present in the ingredients. The amount of water is about 1 % by weight of the total weight of the paint.

15     The compositions of the invention may also include other commonly used paint ingredients, such as pigments, fillers, additives such as tixotropic substances, softeners, solvents, et cetera, to achieve the properties desired by the user.

20     Following advantages, for instance, are achieved by using epoxy silane instead of organo-oxysilane in the composition of the invention:

25     - epoxy silane has properties improving paint adhesion,  
          - epoxy silane acts as a crosslinking agent between the epoxy and siloxane chains,

30     - when using epoxy silane in a siloxane reaction, the amount of alcohol liberated in the reaction in relation to the molecular weight is considerably lower than when using organo-oxysilane. Consequently, a higher dry solids content and a lower amount of volatile organic compounds (VOC) can be achieved.

35     - epoxy silane reduces the viscosity of the paint,  
          - epoxy silane also has a modifying effect on functional fillers, such as wollastonite, whereby strength and anti-corrosive properties improve.

The following examples illustrate the invention.

30     **Example 1 (Paint A, reference)**

Preparation of a polyurethane paint

35     A resin and pigment blend was prepared by first combining 200 g of hydroxyl polyacrylate (Desmophen 365<sup>TM</sup>, equivalent weight 607, manufactured by Bayer) and 10 g of bentonite-based thickener (Bentone 38<sup>TM</sup>, manufactured by Rheox) and blending for about 10 minutes, whereafter 250 g of

titanium dioxide pigment and 25 g of barium sulphate filler were added. The mixture was ground to 20 µm fineness of grind at a dispersing temperature of about 50 °C. The paint was completed by adding the rest of the binder (200 g) and 70 g of butyl acetate and 87 g of xylene to achieve the desired viscosity.

5 The hardener was prepared by mixing 84.65 g of polyisocyanate (Desmodur N 75™, equivalent weight 255) and 15.30 g of butyl acetate.

The specific weight of the paint was 1.31 kg/dm³ (1.01 equivalents per dm³) and the specific weight of the hardener was 1.04 kg/dm³ (3.47 equivalents per dm³).

10

#### Example 2 (Paint B, reference)

##### Preparation of an epoxy paint

A resin and pigment blend was prepared by combining 307.5 g of liquid Bisphenol-A epoxy resin (Dow DER 331™, equivalent weight 190), 34 g of benzoic alcohol and 34 g of xylene to achieve the desired viscosity and 8.5 g of polyamide wax thickener (Crayvallac Super™). After mixing the blend for about 10 minutes, 134 g of titanium dioxide pigment and 278.5 g of feldspar filler (Siokal FF 30™) were added. Thereafter the mixture was dispersed with a laboratory dissolver at a temperature of 55 °C where it was allowed to stay for 15 to 20 minutes. The required fineness of grind was 50 µm. The paint was completed by adding the solvents: 21.5 g of butanol and 33 g of xylene.

15 The hardener was prepared by combining 188.5 g of hydrocarbon resin (Necires EXP-L™) and 4.9 g of polyamide wax thickener (Crayvallac Super™). The blend was allowed to mix for about 10 minutes, whereafter 184.5 g of isophoron diamine epoxy adduct, 250 g of talcum and 58 g of xylene were added. Thereafter, the blend was dispersed with a laboratory dissolver at a temperature of 55 °C where it was allowed to stay for 15 to 20 minutes. The required fineness of grind was 50 µm. The hardener was completed by adding 20 the solvents: 25 g of butanol and 51 g of xylene.

25 The specific weight of the paint was 1.55 kg/dm³ (2.94 equivalents per dm³) and the specific weight of the hardener was 1.29 kg/dm³ (2.85 equivalents per dm³).

**Example 3 (Paint C, present invention)**  
**Preparation of an epoxy polysiloxane paint**

A resin and pigment blend was prepared by combining 306 g of methoxy-functional polysiloxane (Dow Corning 3074™) and 21.3 g of polyamide wax thickener (Crayvallac Super™). After allowing the blend to mix, 156 g of titanium dioxide pigment, 30 g of talcum, 54.5 g of wollastonite, 49 g of feldspar filler (Siokal FF 30™) were added. Thereafter the blend was dispersed with a laboratory dissolver at a temperature of 65 °C. The required fineness of grind was 40 µm. The paint was completed by adding 50.6 g of glycidoxypropyltrimethoxysilane (Silquest A-187™) and 268.5 g of pentaerythritepolyglycidylether (Polypox R 16™).

The hardener was prepared by combining 173 g of polyamide (Versamid 140™) and 33.9 g of aliphatic epoxy resin (Dow DER 732™, equivalent weight 320). The obtained adduct was allowed to react for 24 hours at room temperature, whereafter 404 g of  $\gamma$ -aminopropyltrimethoxysilane (Silquest A-1100™) and 16.2 g of tin catalyst (DBTL) were added.

The specific weight of the paint was 1.46 kg/dm<sup>3</sup> (2.80 equivalents per dm<sup>3</sup>) and the specific weight of the hardener was 0.97 kg/dm<sup>3</sup> (8.19 equivalents per dm<sup>3</sup>).

**Example 4 (Paint D, present invention)**  
**Preparation of an epoxy polysiloxane paint**

Example 3 was repeated, except that 52.7 g of  $\beta$ -(3,4-epoxycyclohexyl)-ethyltrimethoxysilane (Silquest A-186™) was used as the epoxy silane.

The hardener was prepared by combining 217 g of polyamide and 43.5 g of aliphatic epoxy resin (Dow DER 732™, equivalent weight 320). The obtained adduct was allowed to react for 24 hours at room temperature, whereafter 381 g of N-( $\beta$ -aminoethyl)- $\gamma$ -aminopropyltrimethoxysilane (Silquest A-1120™) and 19.8 g of tin catalyst were added.

The specific weight of the paint was 1.46 kg/dm<sup>3</sup> (2.79 equivalents per dm<sup>3</sup>) and the specific weight of the hardener was 1.02 kg/dm<sup>3</sup> (11.17 equivalents per dm<sup>3</sup>).

The specific weight of the paint was 1.46 kg/dm<sup>3</sup> (2.79 equivalents per dm<sup>3</sup>) and the specific weight of the hardener was 1.02 kg/dm<sup>3</sup> (11.17 equivalents per dm<sup>3</sup>).

## 5 Weather resistance

In an ultraviolet tolerance test, the weather resistance of polyurethane paint A, epoxy paint B and epoxy polysiloxane paints C and D were compared.

10 The test was conducted with a QUV/spray-type test equipment. The lamps used were lamps transmitting long-wave ultraviolet radiation (UVA 340 nm). The equipment was run as follows: 4 h UV (T=60°C), 4 h condensation (T=40 °C). The test results are presented in the attached Figure 1.

15 The obtained results show that the epoxy polysiloxane paint has improved resistance to ultraviolet light as compared with the conventional epoxy paint. An average 50 % gloss reduction occurred in 640 hours in the epoxy polysiloxane paints, whereas in the epoxy paints it occurred as early as in 120 hours from the beginning of the test. Gloss retention was clearly best in the polyurethane paint.

20

## Adhesion in corrosion stress

Epoxy paint B and epoxy polysiloxane paint C were exposed to a neutral salt fog test according to SFS 3707 standard. Paint adhesion to a substrate before and after the test indicates the protection performance of the system in extremely severe conditions (pre-treatment: Sa 2½, sheet thickness 5 mm). The test results are presented in Table 1.

Table 1

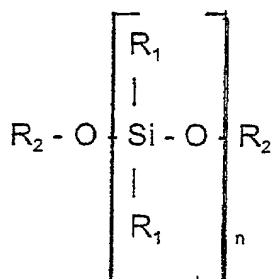
30

Paint	Film thickness (µm)	Tensile value (MPa) before test	Tensile value (MPa) after test
C	120	14.3	7.3
B	200	12.3	3.5

## CLAIMS

1. A composition to be used in paints, characterized by comprising a resin constituent which includes

- i) a non-aromatic epoxy resin,
- 5 ii) a polysiloxane having the formula:



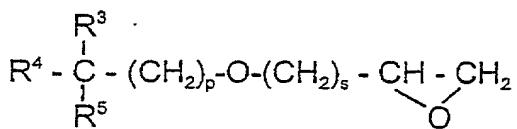
10 where  $\text{R}^1$  is a hydroxyl or an alkyl, aryl or alkoxy group having up to 6 carbon atoms,  $\text{R}^2$  is a hydrogen or an alkyl or aryl group having up to 6 carbon atoms and  $n$  is a number selected so that the molar mass of the polysiloxane is within the range of 400 to 2000, and

15 iii) an epoxy silane which acts as a crosslinking agent between the epoxy and siloxane chains.

20 2. A composition as claimed in claim 1, characterized in that the weight ratio between the epoxy silane, polysiloxane and non-aromatic epoxy resin is 1:2-5:2-5.

25 3. A composition as claimed in claim 1 or 2, characterized in that the non-aromatic epoxy resin is a branched aliphatic epoxy resin.

4. A composition as claimed in claim 3, characterized in 30 that the aliphatic epoxy resin has the formula

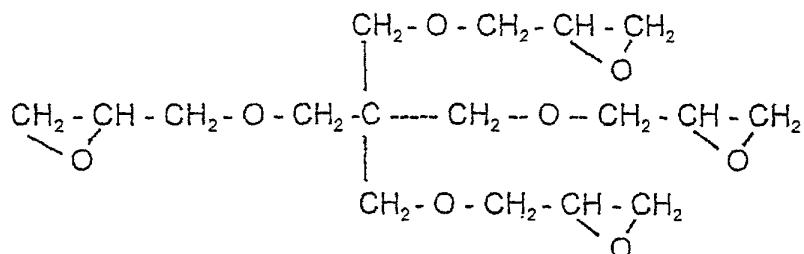


where  $p$  is an integer between 0 and 3,  $s$  is an integer between 1 and 3,  $\text{R}^3$  and  $\text{R}^4$  represent independently  $\text{C}_{1-6}$ alkyl or a group  $-(\text{CH}_2)_p - \text{O} - (\text{CH}_2)_s - \text{CH} - \text{CH}_2$ ,

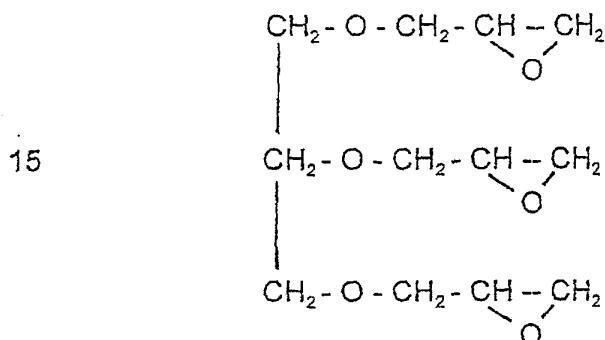
35 where  $p$  and  $s$  are as defined above and  $\text{R}^5$  is hydrogen,  $\text{C}_{1-6}$ alkyl or a group  $-(\text{CH}_2)_p - \text{O} - (\text{CH}_2)_s - \text{CH} - \text{CH}_2$ , where  $p$  and  $s$  are as defined above.

11

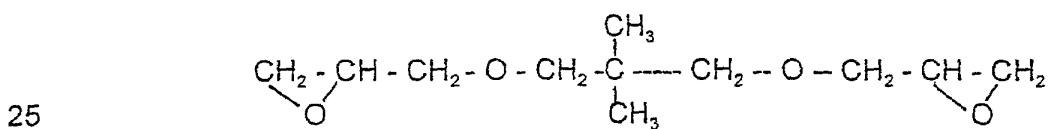
5. A composition as claimed in claim 4, characterized in that the aliphatic epoxy resin has the formula



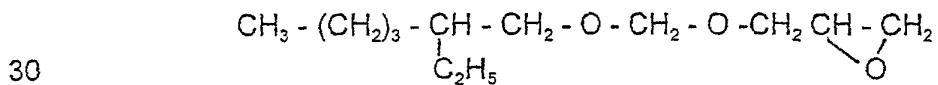
6. A composition as claimed in claim 4, characterized in 10 that the aliphatic epoxy resin has the formula



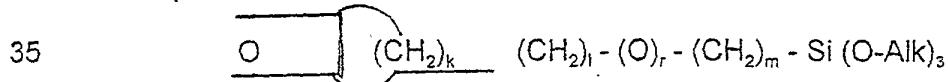
20. 7. A composition as claimed in claim 4, characterized in that the aliphatic epoxy resin has the formula



8. A composition as claimed in claim 4, characterized in that the aliphatic epoxy resin has the formula



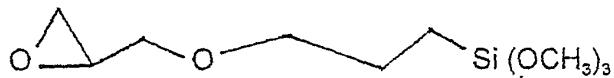
9. A composition as claimed in any one of claims 1 to 8, characterized in that the epoxy silane has the formula



12

where k is an integer between 0 and 4, r is 0 or 1, l is an integer between 1 and 6, m is an integer between 1 and 6 and Alk is an alkyl group having 1 to 6 carbon atoms.

10. A paint composition as claimed in claim 9, **characterized**  
5 **in that** the epoxy silane has the formula



11. A paint composition as claimed in claim 9, **characterized**  
10 **in that** the epoxy silane has the formula



12. A kit, **characterized** by comprising a container A, which  
15 contains a composition according to any one of claims 1 to 11, and a container  
B, which contains a hardener, whereby the container A and/or B may further  
contain conventional additives.

10/019962

1/1

CHANGE IN GLOSS IN QUV TEST (UVA 340 nm)

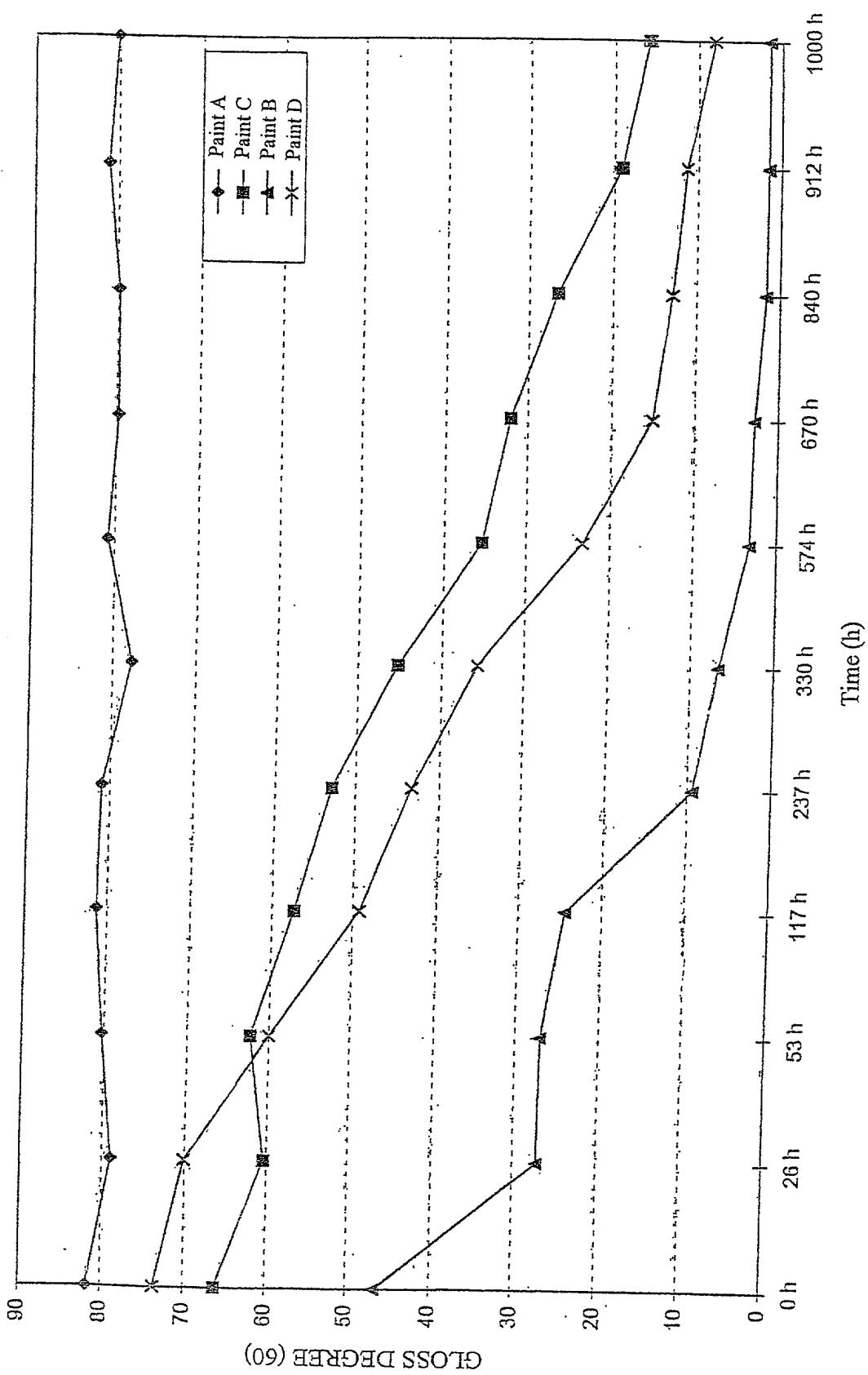


Figure 1

## **COMBINED DECLARATION AND POWER OF ATTORNEY**

As a below named inventor, I hereby declare that

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

## **COMPOSITION TO BE USED IN PAINTS**

the specification of which: *(check one)*

## **REGULAR OR DESIGN APPLICATION**

[ ] is attached hereto.

[ ] was filed on as application Serial No. and was amended on (if applicable).

**PCT FILED APPLICATION ENTERING NATIONAL STAGE**

[ X ] was described and claimed in International application No. PCT/FI00/00613 filed on 4 July 2000 and as amended on (if any).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, '1.56.

**PRIORITY CLAIM**

I hereby claim foreign priority benefits under 35 USC 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed.

**PRIOR FOREIGN APPLICATION(S)**

Country	Application Number	Date of Filing (day, month, year)	Priority Claimed
Finland	991535	5 July 1999	Yes

*(Complete this part only if this is a continuing application.)*

I hereby claim the benefit under 35 USC 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of 35 USC 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations '156 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)

(Filing Date)

(Status--patented, pending, abandoned)

## POWER OF ATTORNEY

The undersigned hereby authorizes the U.S. attorney or agent named herein to accept and follow instructions from as to any action to be taken in the Patent and Trademark Office regarding this application without direct communication between the U.S. attorney or agent and the undersigned. In the event of a change in the persons from whom instructions may be taken, the U.S. attorney or agent named herein will be so notified by the undersigned.

As a named inventor, I hereby appoint the registered patent attorneys represented by Customer No. **000466** to prosecute this application and transact all business in the Patent and Trademark Office connected therewith, including: Robert J. PATCH, Reg. No. 17,355, Andrew J. PATCH, Reg. No. 32,925, Robert F. HARGEST, Reg. No. 25,590, Benoît CASTEL, Reg. No. 35,041, Eric JENSEN, Reg. No. 37,855, Thomas W. PERKINS, Reg. No. 33,027, and Roland E. LONG, Jr., Reg. No. 41,949,

c/o YOUNG & THOMPSON,  
Second Floor,  
745 South 23rd Street,  
Arlington, Virginia 22202.



Address all telephone calls to Young & Thompson at 703/521-2297. Telefax: 703/685-0573.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or first inventor: Mika PERÄLÄ | - 00  
(given name, family name)

Inventor's signature Mika Perälä Date 21 December 2001

Residence: Lahti, Finland PIX Citizenship: Finnish

Post Office Address: Esikonkatu 6 as. 10, FIN-15240 Lahti, Finland

Full name of second joint inventor, if any: Seppo TIKKANEN | - 00  
(given name, family name)

Inventor's signature Seppo Tikkonen Date 21 December 2001

Residence: Järvenpää, Finland PIX Citizenship: Finnish

Post Office Address: Purjopolku 6, FIN-04400 Järvenpää, Finland

Full name of third joint inventor, if any:  
(given name, family name)

Inventor's signature \_\_\_\_\_ Date \_\_\_\_\_

Residence: \_\_\_\_\_ Citizenship: \_\_\_\_\_

Post Office Address: